Ecological transfer of radionuclides and heavy metals at NORM and TENORM sites in Norway

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Abstract: Fen Central Complex (FCC) in southern Norway is an area with thorium (^{232}Th) rich carbonatite rocks. Mining for iron (Fe) and niobium (Nb), also present in abundance in these rocks, was conducted during past centuries. As a result today, technologically enhanced naturally occurring radioactive materials (TENORM) are situated in the area, together with naturally occurring radioactive materials (NORM). Analysis of different environmental samples from FCC showed increased concentrations of ^{232}Th, uranium (^{238}U) and heavy metals – arsenic (As), lead (Pb) and cadmium (Cd). In order to see ecological transfer of both radionuclides and heavy metals, the concentrations in typical Fen vegetation and earthworms species were analyzed. Obtained transfer factors for ^{232}Th were in range from 3 x 10^{-5} to 3 x 10^{-2} and from 3 x 10^{-3} to 9 x 10^{-2}, for vegetation and earthworms, respectively. Similarly, transfer factors for ^{238}U were in range from 9 x 10^{-4} to 4 x 10^{-2} for vegetation and from 3 x 10^{-3} to 7 x 10^{-1} for earthworms. The highest uptake of ^{232}Th and ^{238}U was seen in moss, lichen and earthworms. Heavy metals concentrations were also enhanced in moss. However, radiological risk assessment with site specific information showed the absorbed dose significantly lower than screening level of 10 µGy h^{-1}, suggesting no risk from investigated radionuclides.

1. INTRODUCTION

Naturally occurring radioactive materials (NORM), i.e., ^{238}U, ^{232}Th and their daughter products, are major contributors to the total exposure dose of ionizing radiation of man. Substantial work has been done in the areas of high NORM and TENORM [1-3]. Need for assessment of radiological risk for non-human biota in various natural and man-made scenarios has been emphasized worldwide. With respect to that, activity concentrations of naturally occurring radionuclides in the environmental media (soil, air, water, sediment) and organisms of interests are of high importance. Additionally, non-human biota is rarely exposed to only radiation, but also often to heavy metals. Thus, it is significant to predict the most probable exposure scenario and to investigate both levels of radionuclides and heavy metals in living organisms, as well as to investigate possible diverse i.e., additive, synergistic or antagonistic biological effects [4].

Fen Central Complex in southern Norway is geologically specific volcanic area of carbonatite rocks rich in NORM (^{232}Th and ^{238}U), but also in Fe, Nb and rare earth elements (REE). This area is well known with its high exposure doses to humans, mainly due to high levels of radon (^{222}Rn) [5]. Several mining sites were exploited in this area for Fe, Nb and scandium (Sc) during the past centuries.

Although mining activities were finished in 1960s, TENORM sites are still important sources of environmental pollution along with NORM sites [5].

The main objective of this study was to investigate soil concentrations of naturally occurring radionuclides ^{232}Th and ^{238}U, and some heavy metals such as As, Pb and Cd, as well as, their uptake in the certain living organism in the area. According to that, transfer factors were calculated using organisms concentration and soil concentration. Assessment of environmental impact considering radionuclides is done on the basis of obtained activity concentrations of radionuclides using the ERICA assessment tool [6]. Further activities will be to investigate other radionuclides (^{232}Th and ^{238}U daughters) that could affect living organisms in the area.

2. MATERIALS AND METHODS

Fen Central Complex, positioned 120 km southeast of Oslo, is an area rich in volcanic carbonatite rocks: Sövite, Rödbergite, Rauhaugit and Fenite. This part of country is known with its high ^{232}Th ore deposits, ^{222}Rn levels and consequently, high annual radiation exposure doses [5].

Collection of soil, vegetation and earthworms were done during four fieldworks, organized in period 2008-2009. Sampling sites were chosen with respect to the measured terrestrial gamma dose rates in the air. Soil was taken from depth 0-20 cm as bulk samples. Earthworms (70 adult organisms) were collected from at least 5 different points at each location, by hand sorting. After identifying in the laboratory, they were washed and starved for 2 days until their gut contents were completely egested. Earthworms were then frozen in liquid nitrogen, freeze dried and ground to a fine powder. Plants, that were found to be the most abundant in the area i.e., lichen, moss, birch, pine and spruce, were collected in paper bags, at least 10 different plants from each species. All plants were, further in the laboratory, cleaned from visible soil, washed several times with distilled water and dry for several weeks at temperature 40°C. They were homogenized by milling. Soil samples were air dried for several weeks, crushed to a fine powder and finally homogenized by sieving through 2 mm sieve. Aliquots of soil were dried at 105°C to constant mass and analyzed for pH, water and organic matter content prior to decomposition for analysis.

Concentrations of radionuclides ^{232}Th, ^{238}U and heavy metals in all samples were measured with ICP-MS of extracts obtained by microwave decomposition (Milestone Inc., Ultraclave High performance reactor, Shelton, CT), employing high-grade purity HNO_3 acid for soil and HNO_3/H_2O_2 for plant and earthworm samples. ICP-MS analysis was done using a Perkin ElmerSciex Elan 6000 (Norwalk CT, USA). Quality control assurance was performed by substantial use of internal standard, method and instrument blanks and proper certified reference materials. Activity concentrations of radionuclides (Bq/kg) were obtained by multiplying concentrations (mg/kg) obtained by ICP-MS with factors 4.06 and 12.35, for ^{232}Th and ^{238}U, respectively.

In addition to the total soil analysis, sequential extraction was conducted on all soil samples [7]. The aim was to determine the different radionuclides and heavy metal phases in the soil, mobility and environmental availability.

3. RESULTS AND DISCUSSION

Total soil analysis showed high concentrations of radionuclides in soil, 11258 ± 370 Bq/kg and 87 ± 60 Bq/kg, of ^{232}Th and ^{238}U, respectively. Obtained values exceed the world average values for soil, 45 Bq/kg ^{232}Th and 33 Bq/kg ^{238}U, given by UNSCEAR [8]. Average values of heavy metals (62 ± 6 mg/kg of As; 155 ± 7 mg/kg of Pb and 2 ± 1 mg/kg of Cd) were approximately 2-3 times higher than non-polluted soil values given by Norwegian Pollution Control Authority [9].

Local contamination of terrain was, the most probably, result of physical and chemical weathering of bedrock coupled with former human mining activities. Intensive non-homogeneous distribution of radionuclides was observed. Presence of increased concentrations of heavy metals along with significantly high concentrations of ^{232}Th and moderately high of ^{238}U could pose an increased risk for non-human biota in term of multiple stressors exposure.

Results of sequential extraction analysis revealed potentially different mobility and bioavailability of investigated elements (Figure 1). High percentages of ^{232}Th (94%) and As (87%) were found to be irreversibly bound in soil crystalline oxides of Fe and Mn. It implicated that both radionuclide and

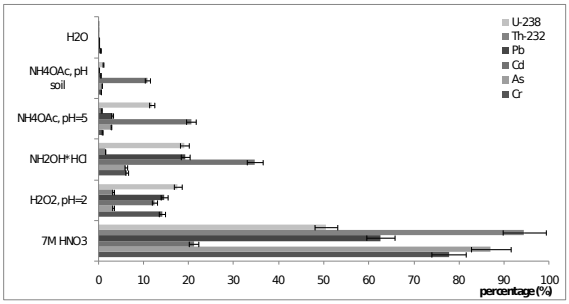

Figure 1. Soil sequential extraction analysis

heavy metal could not be readily taken up by vegetation in the area. However, ^{238}U, Pb and Cd were found to be reversibly bound in soil fractions (50% of ^{238}U, 21% of Cd and 62% of Pb was found to be fixed in residual soil fraction) and could be mobilized and further transported with change of environmental conditions.

Uptake of radionuclides and heavy metals by vegetation and earthworms was studied using transfer factors (TF), obtained as ratio between dry weight plant concentration and dry weight soil concentration (Table 2). Although the vegetation tissue concentrations were estimated as significant, results for transfer factors for ^{232}Th were 1–2 orders of magnitude lower than values previously published worldwide [1, 2, 10-12] ^{238} U transfer factors were, similarly, lower than those obtained in same studies. These results were most likely because of considerably high total soil radionuclide concentrations, used in calculations. To obtain the more accurate information on biological uptake and bioconcentration, the assessment with available soil radionuclide concentrations will be evaluated in future. Moss

and lichen showed, in general, the highest uptake level of both radionuclides and heavy metals. Bioconcentration of ^{232}Th was one magnitude of order lower than of ^{238}U, what was expected since ^{238}U is more soluble and might mimic essential elements.

Table 2. Transfer factors (TF) for investigated radionuclides and heavy metals

Sample	^{232}Th	^{238}U	As	Cd	Pb
moss	2 x 10^{-3}	4 x 10^{-2}	5 x 10^{-2}	2 x 10^{-1}	1 x 10^{-1}
spruce	3 x 10^{-5}	9 x 10^{-4}	5 x 10^{-4}	2 x 10^{-2}	9 x 10^{-4}
fern	3 x 10^{-5}	1 x 10^{-3}	6 x 10^{-4}	4 x 10^{-2}	6 x 10^{-4}
lichen	2 x 10^{-3}	3 x 10^{-2}	5 x 10^{-3}	4 x 10^{-2}	2 x 10^{-2}
pine	7 x 10^{-5}	3 x 10^{-3}	8 x 10^{-4}	3 x 10^{-2}	2 x 10^{-3}
earthworms	8 x 10^{-2}	3 x 10^{-1}	5 x 10^{-1}	11.77	3 x 10^{-1}

In comparison to analyzed vegetation, earthworms showed somewhat enhanced bioconcentration regarding radionuclides, 8 x 10^{-2} and 3 x 10^{-1}, for ^{232}Th and ^{238}U, respectively (Table 2). Few data about transfer of ^{232}Th in natural forest conditions (on contrary to experimentally designed) were found by the authors of this paper. The main source of uncertainty in radiological risk prediction for non-human biota is the value of the transfer parameters used, so developing the available transfer databases should be a focus of future research efforts [13]. In light of these facts, results from this study could be of importance.

TFs for ^{238}U and heavy metals were approximately three times higher than for ^{232}Th, what reflected chemical nature of both elements and availability revealed by soil sequential results. No linearity between TF or log TF and the soil concentration was found for ^{232}Th and ^{238}U. Transfer factors for investigated heavy metals were similar to results published for non-contaminated soil, indicating low bioaccumulation and no increased risk in population term.

In general, the obtained transfer results suggested enhanced uptake of ^{232}Th and moderate uptake of ^{238}U. Thus, the risk for biomagnification of the investigated radionuclides through the food chain could be possible. However, the levels of heavy metal uptake did not indicate increased accumulation in earthworms.

The total dose rates and radiological risk were preliminary obtained by the ERICA assessment tool [6] on the basis of media concentrations of selected radionuclides (Figure 2).

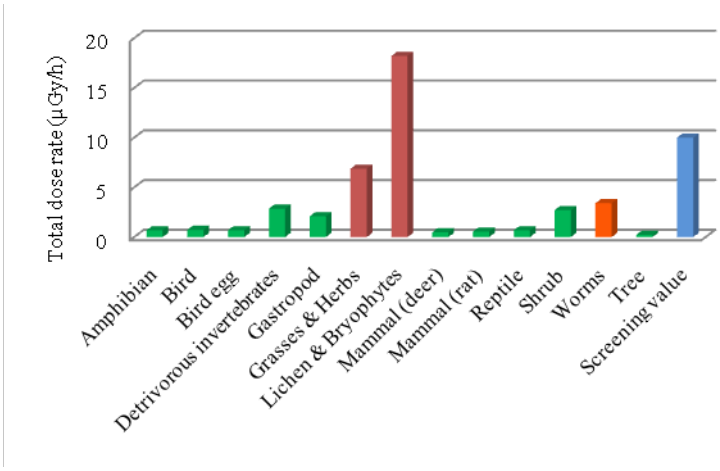

Figure 2. Total exposure dose per terrestrial organism groups

Lichen and bryophytes, grasses and herbs, as well as potentially earthworms were estimated to be the most sensitive and vulnerable organisms in this area (Figure 2). However, detailed assessment, done for lichen with site specific data (activity concentrations and CR value of organism), showed total absorbed dose of 0.58 µGy h^{-1}, significantly lower than screening level (10 µGy h^{-1}) and than value obtained in preliminary ERICA tool estimation(16 µGy h^{-1}). This result reflected, most likely, relatively low uptake in lichen and showed the need to include available site specific data in risk assessment of each organisms group.

Furthermore, although obtained result unequivocally suggested radiation risk lower than unity, it should be emphasized that calculated risk is only from ^{232}Th and ^{238}U. Thus, to obtain complete information on non-human biota radiation doses in the area, daughter radionuclides (e.g., Ra-, Po- and Pb-isotopes) should be included into assessment.

4. CONCLUSIONS

The main objective of this study was to investigate soil concentrations of naturally occurring radionuclides ^{232}Th and ^{238}U and some heavy metals such as As, Pb, Cd and their uptake in living non-human organisms in the Fen area, Norway. Data on the activity concentration of naturally occurring radioactive materials in the environmental media and organisms of interests are of high importance for assessment of radiation impact on non-human biota.

Analysis of Fen soil samples showed high concentrations of both ^{232}Th and ^{238}U, significantly higher than reference values given by UNSCEAR [8]. The found radionuclides concentrations in the soil suggested very inhomogeneous distribution and potential various influence on living organisms. Presence of enhanced levels of As, Pb and Cd suggested multiple stressors scenario in the area. Sequential extraction showed that ^{232}Th and As were rather immobile, irreversibly fixed in

crystalline soil fractions, while ^{238}U, Pb and Cd were distributed in soil in such way that changing of environmental conditions could mobilize them and make them bioavailable.

Concentrations of ^{232}Th and ^{238}U in moss and lichen were higher than those found worldwide [1, 2]. However, no much data for comparison purposes is available on wild species grown in natural forest rich in ^{232}Th. Although high levels of radionuclides were found in some plants, obtained transfer factors were approximately one order of magnitude lower that those published worldwide previously. The explanation could be in limited plant uptake of radionuclides in conditions of extremely high soil concentrations of ^{232}Th and moderately high of ^{238}U. To obtain the better insight on vegetation ability to concentrate radionuclides, the available transfer factors (ATF) will be evaluated in future. Concentrations and transfer factors obtained for earthworms were, on contrary, higher than those published by ERICA [6], indicating the active biological uptake by these organisms. However, radiological risk assessment with site specific information showed the absorbed dose significantly lower than screening level of 10 μGy h^{-1}, suggesting no risk from investigated radionuclides.

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